Electron tunneling into epitaxial films of $Nd_{2-x}Ce_xCuO_{4-y}$

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We have performed tunneling experiments on planar junctions $Al/AlO_x/Nd_{2-x}Ce_xCuO_{4-y}$. This is the first experiment using epitaxial films with the planar junction for electron-doped high T_c $Nd_{2-x}Ce_xCuO_{4-y}$. The temperature dependencies of the gap features were measured, in which the phonon structures were clearly observed in the normalized conductance curve. These results show accord with the phonon peaks in the phonon density of states obtained recently by neutron inelastic scattering experiments.

The tunneling experiments serve as a useful technique to obtain information about gap features and/or phonon densities of the states of superconductors.[1] In addition, these can provide insight into the mechanism responsible for electron pairing. For these reasons, a great deal of tunneling experiments have been performed on high T_c oxide superconductors (HTSC). There are, however, still unclear points about the pairing mechanism and whether the HTSC has the BCS-like energy gap. The main difficulty in the tunneling experiments for the HTSCs is due to its short coherence length; namely, it is shorter than the distance between Cu-O planes. In the case of $Nd_{2-x}Ce_xCuO_{4-y}$ (NCCO),[2] the coherence length ξ along the c axis is 3.4 Å (Ref. 3) and the distance between the Cu-O planes is $\sim$6.06 Å. Since the tunneling proves the region from the surface close to the coherence length, the results are susceptible to the surface conditions.

In this letter, we report the results of tunneling experiments on planar junctions of the NCCO epitaxial films prepared by the method of molecular beam epitaxy (MBE). The results of $(dI/dV)-V$, $(-d^2I/dV^2)-V$ curve show clearly fine structures, and some peaks are in accord with the phonon peaks in the phonon density of states obtained from neutron inelastic scattering experiments.[4] In addition, there are in accord with the peaks of $\alpha^2F(\omega)$ derived from the point-contact tunneling experiments on polycrystalline NCCO by Huang *et al.*[5] We also found specific features on NCCO, such as the existence of large zero-bias conductance, linear background conductance, and gap broadening.

The NCCO epitaxial films (1000 Å) were prepared on the $SrTiO_3$ (100) substrate by MBE. Knudsen cells were used for the sources of Nd, Cu, and Ce. Each of the deposition rates was monitored by the quartz crystal oscillator method throughout the deposition. Oxygen gas was flowed toward the substrate from the pinholes of the pipe located beneath the substrate, and the typical oxygen pressure was 10^{-4} Torr around the substrate. To oxidize fully, the films were post-annealed in air at 900 °C for 2 h and then quenched to room temperature. Reduction treatment, which introduces oxygen deficiency ($y\sim0.04$), is necessary in order to obtain superconductivity in the case of NCCO.[6] These films were reduced in an Ar atmosphere (0.4 Torr, with O_2 pressure less than 10^{-6} Torr) at a temperature from 550 to 700 °C for 15–20 min. It should be noted that this reduction is extremely sensitive to time, pressure, temperature, and film thickness.[7] X-ray analysis shows only sharp (001) peaks, indicating the films were grown with the axis preferentially oriented normal to the substrate.

The junctions were fabricated in the following manner: first, Al was deposited on the NCCO films and then oxidized in air for several minutes. The thickness of the insulator is approximately 30 Å, which was extrapolated from the surface roughness measurements. The Al film was, again, deposited on top, with the thickness of approximately 800 Å as counterelectrodes. Typical junction resistance was between 30 and 100 Ω and the area was 0.25 $\times$0.5 mm.[2] Since the planar junctions have a lower current density than that of the point-contact method or the STM, the planar junctions are free from higher current density, causing local heating. The additional features are that it is not difficult to observe the fine structures in the conductance curves because of its high stability and reproducibility. The measurements of differential resistance were performed by the standard ac modulation technique in the temperature range from 1.5 to 77 K. The differential conductance was obtained by inverting these differential resistances.

In a BCS superconductor (not restricted to the phonon-mediated pairing mechanism), the quasiparticle density of states at $T=0$ is given by the normalized conductance

$$\left(\frac{dI}{dV}\right)_s \Big/ \left(\frac{dI}{dV}\right)_n = \Re\left(\frac{E}{(E^2-\Delta^2)^{1/2}}\right). \qquad (1)$$

As is well known, in the case of strong coupling superconductors, a phonon structure (or excitations relevant to pairings) emerges in the normalized conductance as the deviation from the BCS density of states.[8] Such structures are usually compared with the phonon density of states obtained from neutron inelastic scattering experiments.

In Fig. 1, we present the observed results of conductances for sample A (planar junction Al/AlO_x/NCCO) in the temperature range from 1.5 to 77 K. Since the T_c onset of the film is 21.6 K, the drastic decrease of the conductance in the vicinity of zero-bias voltage apparently reflects the presence of the energy gap. In addition, the small humps in the observed conductance have appeared at 7, 4.2, and 1.5 K. Our results indicate larger gap broadening than the expected thermal broadening (0.09 meV at 1 K).

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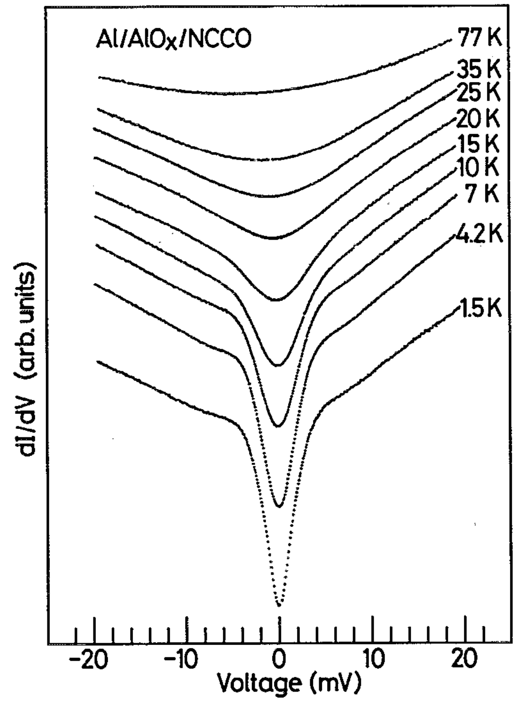

FIG. 1. Temperature dependence of dI/dV of sample A, planar junction $Al/AlO_x/Nd_{2-x}Ce_x\,CuO_4$ from 1.5 to 77 K up to 20 mV. The measured voltage is the Al voltage relative to the NCCO film.

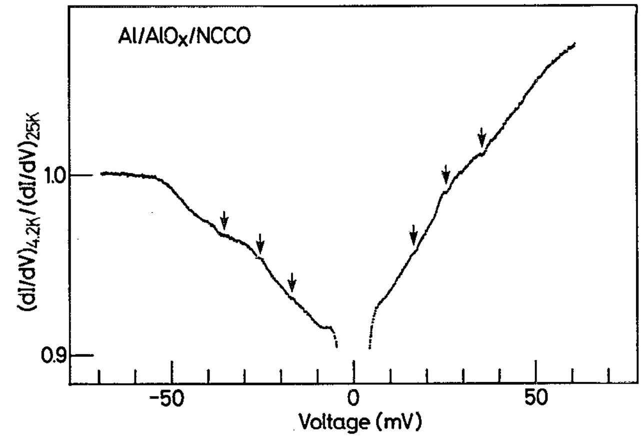

FIG. 2. The normalized conductance $[dI/dV]_{4.2\ K}/[dI/dV]_{25\ K}$ of sample B up to 70 mV. Arrows indicate the structures observed at the identical voltage both in positive and in negative voltage. The measured voltage is the Al voltage relative to the NCCO film.

Dynes *et al.*[9] introduced a parameter Γ into the quasiparticle density of states, replacing E in Eq. (1) by $E-i\Gamma$,

$$D(E,\Gamma)=\Re\{E-i\Gamma/[(E-i\Gamma)^2-\Delta^2]^{1/2}\}. \qquad (2)$$

This phenomenological broadening originates from the effect of the quasiparticle lifetime due to inelastic scattering. The best fitting for the normalized conductance $[dI/dV]_{1.5\ K}/[dI/dV]_{25\ K}$ (not shown here) presents the value of $\Delta=1.5$ meV and $\Gamma=3.1$ meV. The anomalously large value of Γ is due to the fact that the value of $[dI/dV]_{1.5\ K}/[dI/dV]_{25\ K\ (\text{at}\ V=0)}$ is finite and approximately 0.75 in our case. Namely, since a planar junction has a much larger area than the point-contact method or the STM, a certain percent of the contact area is probably in the normal state due to surface inhomogeneity. As a result, the tunneling current through the normal state region enhances the conductance in the vicinity of zero-bias voltage. We have estimated the value of $2\Delta/k_BT_c\sim3.6$ using T_c endpoint of 9.6 K and $\Delta=1.5$ meV, since the film used in our experiments showed a broad superconducting transition. Although this value is quite close to the BCS value 3.53, it involves ambiguity causing from the definition of T_c and Δ.

Linear background conductance was observed even up to 35 K. The explanation given by Varma *et al.*[10] is based on the quasiparticle density of states itself, in which the lifetime of the quasiparticle is proportional to the inverse of $|E-i\Gamma|$, i.e., the density of states has the linear energy dependence. However, it is not consistent with the results on the BiSrCaCuO system, not yielding the linear background conductance.[11] In this connection, several explanations have been proposed, including the inelastic scattering effect,[12] the barrier height effect, and the charging effect.[13]

Figure 2 shows the result of the normalized conductance $[dI/dV]_{4.2\ K}/[dI/dV]_{25\ K}$ of sample B with the voltage up to 70 mV. We have chosen the conductance of negative voltage as unity since it seems to converge to unity more rapidly than that of positive voltage. The normalized conductance shows some unusual features. First, there is no conductance overshoot and, in the negative voltage region, the value of the whole curvature is less than unity, indicating the slope of the normal state conductance is different from that of the superconducting state. At this point, we have no explanation for this but the possibility that the nature of the tunneling barrier changed after the measurement of the superconducting state conductance. Second, asymmetry was observed in the conductance, i.e., the tunneling current from Al to NCCO is larger than that of the opposite direction. These characteristics have also been observed for the case of hole carrier HTSC. Kirtley[13] has given the explanation on this based on the hole-type carrier. Since it is reported that the carrier of NCCO is the electron,[2,6] our results indicate that this is common for both cases of hole and electron carrier HTSC.

The most striking feature of Fig. 2 is the observation of the structures in the normalized conductance at the same voltage in the positive and negative direction. To clarify this point, we have obtained the second derivative $(-d^2I/dV^2)$, as shown in Fig. 3, in which the abscissa shows the energy, $V-\Delta$. Since the normalized conductance of sample B has no sharp peaks in it, the determination of the energy gap by fitting Dynes's equation is not appropriate. For this reason, we assumed that the gap value of sample B is 1.5 meV because samples A and B were fabricated on the identical film. The peaks in $(-d^2I/dV^2)$, which are large and have the same energy in both positive and negative voltage, are 15.0, 23.5, and 33.5 meV. The peaks at 15.0 and 33.5 meV show good agreement with the phonon density of states $F(\omega)$ (short dashed line shown in Fig. 3) obtained from the neutron inelastic scattering experiments by Tralshawara *et al.*[4] and with $\alpha^2F(\omega)$ derived from the previous tunneling experiment by Huang *et al.*[5] The peaks

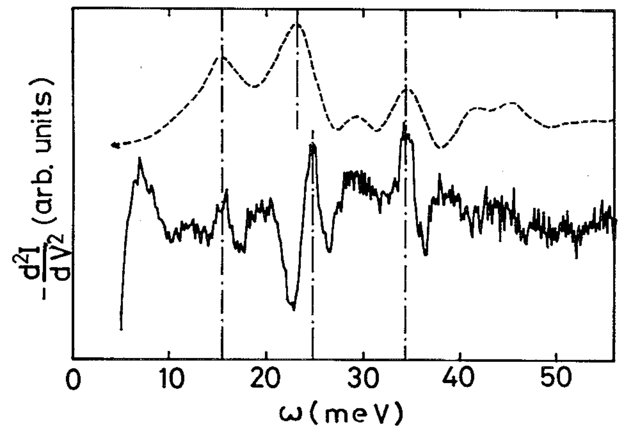

FIG. 3. $(-d^2I/dV^2)$ of sample B vs energy. The peaks are observed at 15.0, 23.5, and 33.5 meV both in the positive voltage direction. The abscissa is energy $\omega(=V-\Delta)$, where $\Delta=1.5$ meV, determined for sample A by fitting the Dynes equation. The short dashed line is the neutron measurements of $F(\omega)$ from Tralshawara *et al.* (Ref. 4).

at 23.5 meV are close to the peak at around 22 meV in $F(\omega)$. In $\alpha^2F(\omega)$ of Huang *et al.* data, junction 1 has the peak at around 20 meV but junction 2 does not. Although the reason for this uncertainty is unknown, obviously it is interpreted that both the lower and the extended higher energy peaks in $F(\omega)$ correspond to the vibrational modes relevant to heavy atoms such as Nd and Ce, and those relevant to the O atom or the Cu-O plane, respectively.

To summarize, we have performed tunneling experiments on planar junctions of NCCO epitaxial films prepared by MBE. The obtained $(-d^2I/dV^2)$ curve have shown peaks at the energy of 15.0, 23.5, and 33.5 meV, which are in accord with the phonon density of states obtained from the neutron inelastic scattering experiment by Tralshawara *et al.*,[4] indicating the relatively strong electron-phonon interaction. It suggests that phonons are playing a role in pairing in the NCCO. However, this does not rule out the possibility that other excitations are also playing a role in the pairing. Clearly, tunneling and neutron data at high energy would be desirable.

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